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Particle radiation damage in high T_c and conventional superconductors

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Abstract : Particle radiation damage in different high temperature superconductors (HTSC) and in earlier or conventional superconductors like the A-15 compounds has been reviewed with an emphasis on our work. Although it has been studied mainly by measuring electrical resistivity, the possibility of further findings from specific heat measurements was established for the first time in our 1995 work on Tl-superconductors. Superconducting critical temperature T_c, is seen to increase due to irradiation for $Bi_2Sr_2CaCu_2O_{8+\delta}$ or Bi-2212 samples with excess oxygen and decrease for Bi-2212 with O-content equal to or less than the optimum, that gives maximum T_c. This increase as well as the decrease of the superconducting critical temperature can be explained from irradiation induced O-displacements and these O-displacements affecting charge-carrier-concentration, that controls T_c. Rise and fall of T_c on irradiation has also been observed for A-15 superconductors and similarly explained on the basis of changes in the density of states at the Fermi level, involved in the simple BCS formula for T_c. Flux-pinning by radiation induced defects and subsequent increase of the critical current density is attractive for superconducting magnets and related applications.

Keywords : Radiation damage, HTSC, A-15 compounds

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1. Introduction

Fast particle radiation damage [1-3] to superconductors must be accurately understood and possibly prevented during utilization of various superconductors in radiation environments. In fact, their use in accelerators and fusion devices has been started in a big way. Particle irradiation is one of the few controlled ways of introducing defects in a solid, here a superconductor. Damage to the structure generally reduces [1-3] the superconducting critical temperature (for T_{c0} to T_c). An increase of superconducting critical temperature in case of certain implantations (like hydrogen implantation [4] at low temperatures in Pd forming PdH) is due to addition of new atoms to the target forming a new material. But in correct radiation damage experiments, as reported here, the experiment is designed to let the fast particles go out

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of the target to avoid implantation. Often there has been a doubt on whether the rarely observed rise in superconducting critical temperature [5, 6] in such a radiation damage investigation has resulted from an unknown factor like wrong measurement.

The present paper will review, mainly from our own He^{2+}-irradiation experiments, the situation for some "high temperature" oxide superconductors and "low temperature" metal-based superconductors. The change, $\Delta T_c = (T_c - T_{c0})$ depends not only on the nature of the particle, its energy and dose but also on T_{c0} or more specifically the unirradiated state of the sample as seen for even conventional superconductors. Less emphasis on the last factor led to some excitement and confusion regarding a few reports of an apparently surprising increase in T_c of various HTSC due to heavy ion and He irradiations. In fact, the unirradiated state of the HTSC sample, particularly its oxygen-content, determine [7] mobile-carrier-concentration or electronic structure of the unirradiated HTSC.

The dependence of the sign of irradiation-induced ΔT_c on the initial electronic structure can be lucidly illustrated not only for the new oxide superconductors, but also for A-15 superconductors, the highest T_c superconductors of the pre-HTSC decades. It must be added that, along with Nb-Ti alloys, A-15 compounds like Nb_3Sn are still the most widely used practical superconductors for large devices.

There have been substantial use in radiation damage of various HTSC, of the two lightest ions [5, 6] to utilize their longer range to probe thicker samples, and of very heavy ions like Ag [8] and Xe [9] for producing ion tracks that pin fluxlines along the tracks Irradiation induced increase of critical current density of a superconductor due to such flux-pinning has great technological and theoretical importance. As these aspects are being adequately covered in various international reviews, these will be just outlined in the present review.

2. Radiation damage in high T_c A-15 and low T_c A-15 superconductors

A-15 compounds are A_3B type compounds, like Nb_3Sn, having a *bcc* sublattice of B-atoms (Sn-atoms). The A-atoms or Nb-atoms form three orthoganal chains distributing two Nb or Nb-like atoms over each face of the cubic sublattice of the Sn or Sn-like atoms. The density of states at E_F [DOS (E_F)] for the better known A-15 compounds like Nb_3Sn, Nb_3Ge and V_3Ga is high, making their T_c high (Table 1) compared to other conventional superconductors. A peak of their DOS falling on E_F makes DOS(E_F) rather high. The simple BCS expression for T_c predicts higher T_c for higher DOS(E_F), and explains the high T_c, observed in many A-15 compounds. However, for A-15 compounds like Mo_3Ge and Mo_3Si, the DOS peak or peaks [10-13] just misses or miss E_F, making their T_c low compared to the superconducting A-15 compounds (Table 1). It can also be shown that the sharpness of the DOS peak decreases with decreasing integrity or continuity of the above-mentioned A-atom or Nb-atom chains. The integrity of the chains and hence T_c can, therefore, be affected by a fast particle irradiation. Radiation damage in A-15 compounds is of great practical importance in view of an increasing use of superconducting magnets (utilizing A-15 superconducting wires in many cases) in radiation environment like particle accelerators. Here, we limit ourselves to (a) some details on decrease of T_c of Nb_3Sn as observed in our 13.6 MeV and other He^{++}-irradiations, and (b) increase [12-13] of T_c of Mo_3Ge and Mo_3Si due to He-ion and S-ion irradiations.

Table 1. Decrease and increase of T_c in different A-15 superconductors due to damage by different energetic ion irradiations

Ion, target, Ref and T_{co}	fluence ($\times$ cm^2)	
13.6 MeV He, Nb_3Sn, [13], 13.8 K	1.82×10^{17}	10.40 K
02.0 MeV He, Nb_3Sn, [14], 18.1 K	3.30×10^{16}	11.10 K
20.0 MeV S, Mo_3Ge, [15] 01.5 K	3.00×10^{15}	06.5 K
20.0 MeV S, Mo_3Ge, [15], 01.5 K	1.00×10^{16}	03.5 K
20.0 MeV S, Mo_3Si, [15], 01.9 K	3.00×10^{15}	06.5 K
20.0 MeV S, Mo_3Si, [15], 01.9 K	1.00×10^{16}	07.7 K

A superconducting wire consisting of many Nb_3Sn filaments (each of about 5 micron diameter) in a Cu-Sn matrix was irradiated in a liquid Nitrogen Cooled Target Holder by a 40 MeV He^{++}-beam from VECC by the authors of Ref. 10 to see qualitatively the dependence of ΔT_c on beam energy. After irradiation, individual Nb_3Sn filaments from different depths (with espect to the beam direction) were picked up using a chemical to remove the bronze (Cu-Sn) natrix. To measure T_c of an individual Nb_3Sn filament (too brittle to withstand the differential contraction on cooling in an ordinary sample holder) in a liquid He cryostat, a Nb-padded sample-holder was to be specially made. Resistance *vs.* temperature plots (Figure 1) showing on-set and mid-point T_c have thus been obtained for the filament I (marked I in Figure 1), taken from zero depth and hence subjected to 40 MeV beam, filament II, from an intermediate depth and hence subjected to a lowered energy beam, and filament III, from larger depth receiving much fewer particles of the beam at still lower energy. An alpha beam of energy much higher than hundreds of keV or 1 MeV shoots past the lattice ions with little of nuclear (or lattice ion) energy loss [10] and a lot of electronic energy loss. The electronic energy loss does not damage Nb_3Sn, a metallic system. So the 40 MeV beam on filament I is causes less degradation

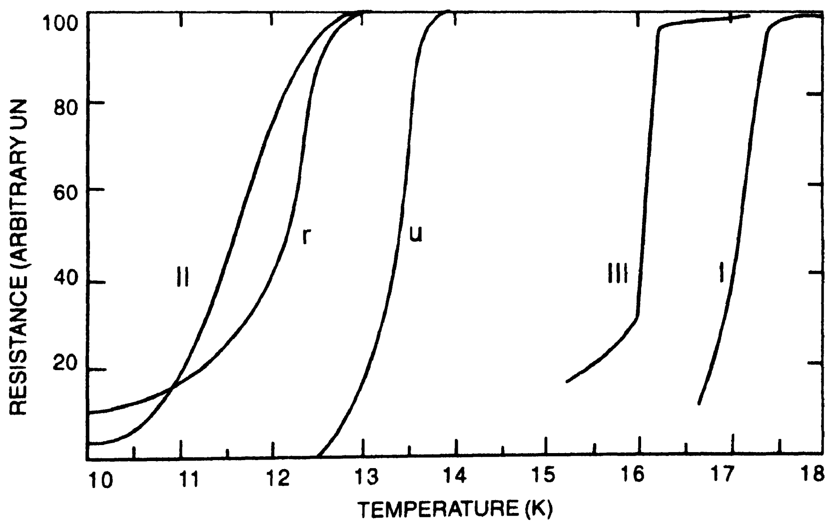

Figure 1. Graphs I, II and III are resistance *vs* temperature plots of individual Nb_3Sn filaments of a Nb_3Sn-in-CuSn MF wire, irradiated by a 40 MeV alpha-beam along a diameter. Filament I is from zero depth, and II and III are from increasingly larger depths. Similar plots for an unirradiated (graph-u) Nb_3Sn layer and after its irradiation by 1.82×10^{17} ions/cm^2 of 13.6 MeV He^{2+}-beam (shown in graph-r).

of T_c than the low energy beam on filament II. References in [10] and TRIM calculations provide rather accurate estimates of these nuclear and electronic energy loss. The nuclear energy loss can be shown to peak at a certain low energy. So beams of still lower energy have lower damaging power. This factor and lowered flux of particles reaching filament III qualitatively explain why its T_c is higher with respect to the filament II.

Above experiment with 40 MeV alphas and our damage energy per atom (dea) calculation [10] showed that impractically long irradiation time will be needed to produce a convincing change of even a few Kelvin in case of the 40 MeV beam. Lower energy implies higher damage till the beam energy is so low (turns out be low sub-MeV values depending on the target-ion pair) as to be unable to transfer at least the displacement energies to the lattice atoms. But to keep the range of the irradiating alphas in Nb_3Sn much longer than 5 micron, the thickness of our [10] Nb_3Sn layer CVD-deposited on a Hastelloy substrate, a beam energy of 13.6 MeV was decided. Still a high dose of 1.82×10^{17} cm^{-2} was needed. Subsequently high target currents of the order of 1 micro-amp. was essential to achieve the high fluence. Beam heating of the order or 13.6 Watt was efficiently removed from the Nb_3Sn target by our design of the target-holder utilizing the large cooling power of water. Thermocouples planted near the sample or target showed the irradiation temperature to be 70° C only.

Above-mentioned 13.6 MeV He^{++}-irradiation dose was chosen to achieve same dea [10] or transfer to the lattice the same mean energy per atom, as the 3.30×10^{16} cm^{-2} dose of 2 MeV He^{++}-irradiation by Poate *et al* [11]. Their experiment on Nb_3Sn with a near ideal T_{c0} of 18.1 K measures (Table 1) $\Delta T_c = -7$ K in contrast to $\Delta T_c = -3.4$ K only by our irradiation to the same dea. This apparent anomaly can be explained from the fact that ΔT_c depends not only on dea and target name, but also on the initial or unirradiated state of the target as given by its T_{c0}. It is noteworthy in Table 1, that irradiations to same dea of samples with different T_{c0} have resulted in more or less same final T_c .

Table 1 next presents observations of positive ΔT_c for two conventional superconductors, positive ΔT_c being seen (in Table 2) also for various HTSC. These increase of T_c on irradiation are for Mo_3Ge and Mo_3Si, A-15 compounds with such electronic structure as to result in low

Table 2. A table of change in T_c and electrical resistivity due to 40 MeV He^{2+}-irradiation Consolidated result for slow-cooled Bi-2212 and (Bi,Pb)-2212 samples under different irradiated conditions giving $T_c = T_c\ (R = 0)$, $T_c^{mf}(\rho)$ = mean field T_c from half the normal resistivity, $T_c^{mf}(p)$ = mean field T_c defined as the position of the peak in the (dR/dT) vs T plot. ρ (300K) = resistivity in m-ohm-cm at 300 K, T_0 = temperature for 2-D to 3-D cross-over as explained in the text, and J = interlayer coupling constant $= [T_0 - T_c^{mf}(p)]/4T_c^{mf}(p)$

He^{++}/cm^2	T_c in K	$T_c^{mf}(\rho)$	$T_c^{mf}(p)$	r (300K)	T_0	1000J
for Bi-2212						
unirrd	64.5	69.1	68.5	007.3	70.7	7 8
2.2×10^{15}	64.0	70.0	69.2	022.2	71.6	8.8
8.14×10^{15}	69.0	73 1	72 0	074 0	74.7	9 2
for (Bi, Pb)-2212						
unirrd.	59.5	64.8	64 0	007.9	65 7	6.6
2.2×10^{15}	63.0	68.3	67.7	064.0	69 6	6.8
8.14×10^{15}	65.0	69.9	70.0	102.5	72.1	7.5

T_{c0} of 1.5 K and 1.9 K only. The sulpnur ion irradiation increases [12, 13] T_c by "improving" $DOS(E_F)$ till the basic structure supporting superconductivity is slowly degraded by higher and higher fluence. The present [12, 13] range of fluence shows (Table 1) this rise and fall for Mo_3Ge, and only the rising part for Mo_3Si.

3. Radiation damage in various HTSC

3.1. Opposite changes in T_c in differently-oxygenated Bi-HTSC due to ion irradiation :

$Bi_2Sr_2CaCu_2O_{8+\delta}$ or Bi-2212 samples have been prepared from a mixture (in such proportion as to give Bi:Sr:Ca:Cu = 2:2:1:2) of Bi_2O_3, $SrCO_3$, $CaCO_3$ and CuO by repeated grinding, pelletizing and prolonged firings in a programmable furnace. After the final firing at 825°C, two types of samples have been prepared. Some of the pellets have been quenched from 825°C to room temperature by collecting the red hot pellets on an aluminium plate directly from the furnace. Other pellets have been slowly cooled (200° C/hour) to let these absorb more oxygen. Powder x-ray diffraction of the pellets showed, Bi-2212 lines [14, 15]. The resistivity *vs.* temperature measurements have been done in a Leybold-Cryogenerator-Setup by the standard four probe method before and after each irradiation. For these measurements, four low resistance contacts (below 0.5 ohm) to the sample have been made by either by ultrasonic soldering or with silver paint. A constant direct current of 300 μA or so from a LakeShore 120 Current Source has been passed through the sample and the drop across the voltage probes measured by a Keithley 182 Nanovoltmeter. The sample temperature has been recorded using a platinum resistance thermometer. The maximum possible error in temperature measurement is estimated to be 0.2K.

Slow-cooled $Bi_2Sr_2CaCu_2O_{8+\delta}$ or Bi-2212 and $(Bi_{0.92}Pb_{0.17})$-2212 pellets have thus been prepared under the same heat treatments and with similar grindings. There has been no other damage studies on (Bi, Pb)-2212, the special role of Pb in tending to expand the Bi-O spacings being important. Due to slow-cooling these samples incorporated excess oxygen.

To let 40 MeV He^{2+}-beam to pass through the samples, each pellet was thinned below 250 micron These irradiations were done in the Variable Energy Cyclotron Centre, Calcutta, using the specially fabricated target-holder. Annular perspex flanges insulated the main body (aluminium) of the target-holder from the cyclotron beamline, to which it was fitted for irradiation. This allowed target-current measurement by a current integrator and an estimate of the fluence. Re-collection of secondary electrons, ejected by the 40 MeV He^{2+}-beam from the target-holder or even the target, by an aluminium cup ensured that He^{2+}-particles alone gave rise to the target-current. Beam-current or target-current higher than 120 nA was never used.

The main results from our measurement of electrical resistivity as a function of temperature and hence $T_c = T_c$ (R=0) are given in Table 2. From excess conductivity analysis, the temperature T_0, above which the 3-dimensional fluctuation changes over to a 2-dimensional fluctuation in the HTS, has been estimated. Estimates of the mean field superconducting transition temperature, T_c^{mf}, has been made in Table 2 by identifying it once to the temperature, $T_c^{mf}(\rho)$, at half the normal resistivity, and then by identifying it to the position, $T_c^{mf}(p)$, of the peak or maximum in (dR/dT) *vs.* T plot. The two definitions are seen to yield comparable values. The latter definition has been used to calculate the interlayer coupling constant :

$$J = [T_0 - T_c^{mf}(p)]/4T_c^{mf}(p).$$

Electrical conduction by holes is known to be limited to individual conducting layers (CuO_2-double-layers) with no conduction in c-direction across the so-called block layers. A block layer consists of a SrO layer and BiO-double-layers. Here, J represents the Josephson coupling that sets in (at and below T_0) among the CuO_2-double-layers through the block layers. Partial substitution of Bi by Pb in the block layers is seen in Table 2 to reduce J as well as T_c. In quenched Bi-2212 with T_{c0} = 79 K, which was checked to be having less than optimum O-content, 40 MeV he^{2+}-irradiation similarly reduced J as well as T_c due to further O knock out by the irradiation. O-displacements by 40 MeV He^{2+}-irradiation on O-rich (and hence with lowered T_{c0}) Bi- and (Bi, Pb)-2212 increases T_c as shown in Table 2. It is satisfying that along with T_c, J is also increased by the irradiation as the sample proceeds towards O-optimization. The direction of change is thus seen to be identical for the interlayer coupling constant and superconducting critical temperature.

3.2. Volume fraction of radiation damage from specific heat measurements :

The main results of our specific heat, C(T) or C, and a.c. susceptibility measurements in the FZ-Karlsruhe set-up [16] on $Tl_2Ba_2Ca_{n-1}Cu_nO_{2n+4}$, $n = 2$ and 3, can be summarized. A jump is expected at T_c in the C/T *vs.* T graph on account of the expected sharp change is the electronic contribution. It is masked by the slowly varying lattice contribution only for HTSC samples, T_c for HTSC samples being a few times higher. This jump could be seen by us in $|(C/T) - (C/T)_{BG}|$ *vs.* T graph where $(C/T)_{BG}$, the background or BG value of (C/T) was estimated by extrapolating (C/T) *vs.* T data in the non-superconducting regions on the two sides of the T_c-region into the T_c-region.

The jump in C/T was seen to be (33 ± 3) mJ mol^{-1} K^{-2} at T_c = 102 K for unirradiated Tl-2212. For unirradiated Tl-2223 it was (28 ± 3) mJ mol^{-1} K^{-2} at T_c = 116 K. Irradiation [2] at VECC with 2×10^{16} of He^{2+} cm^{-2} at 40 MeV reduced this specific heat jump to (i) an unmeasurably low value for Tl-2212 and (ii) to (18 ± 2) mJ mol^{-1} K^{-2} at T_c = 113 K for Tl=2223. Let us note that the size of the (C/T)-jump should indicate the superconducting volume fraction and its position the T_c (if the jump has the ideal saw-tooth shape) or an average T_c (if the shape of the jump is non-ideal).

So, it is clear that our alpha irradiation reduced the superconducting volume fraction significantly (by a factor of 28/18) for Tl-2223 and practically to zero for Tl-2212. However, the onset T_c as measured as our a.c. susceptibility measurement was practically unchanged by this irradiation – a stark contrast to the significant to drastic change in the specific heat signal. This can be explained. It indicates that some traces of the original superconducting phase are left undamaged in each sample. These traces are enough to provide shielding currents and thus show almost unchanged onset T_c in the a.c. susceptibility measurement. So, a more realistic probing of the damaged bulk of the sample can be achieved [2] from the specific heat measurement.

Our observation of more damage to Tl-2212 than to Tl-2223 provides further hints into the details of the damage process. It can be explained from the fact that Tl-2212 consisting of TlO-BaO-CuO-Ca-CuO-BaO-TlO layers has, in comparison to Tl-2223 or TlO-BaO-CuO-Ca-CuO-Ca-CuO-BaO-TlO, has relatively fewer conducting layers (metal-like Cu-O or rather CuO_2 layers) and relatively more blocking or non-conducting layers. Here one has the evidence that damage to these oxides by the high energy (40 MeV) alpha-beam must be mostly due to electronic energy loss [9, 10], a practically non-damaging process in metals and alloys. Metal-like CuO_2 layers, relatively more in Tl-2223, are less affected by electronic energy loss and hence by our 40 MeV alpha-irradiation. It, however, affects the blocking layers rather effectively. Hence, a larger volume fraction is affected in Tl-2212 that has relatively more blocking layers.

3.3. Beneficial effects of radiation damage to the critical current density :

Very heavy ions like Xe and Ag with even 5 MeV per nucleon pass practically undeviated in common solids including the HTSC [3, 8, 9, 16] to generate straight ion tracks. Such tracks in a high T_c sample are practically non-superconducting, and offer, under certain matching conditions, lower potential energy sites or "pinning sites or centres" for magnetic fluxlines, that thread the (type 2) superconductor in a magnetic field. If suitable ion tracks of suitable number density (matching the magnetic flux density to be faced by the HTSC) are generated by the irradiation in the c-direction of the HTSC samples, the necessary fluxpinning can be achieved to increase the a-b plane current density of the HTSC. Inspiring increase of the current density by factors up to 5 in zero external field and by factors up to 4 in an external field of 3 T have been reported in one of the studies. To generate more and more pinning tracks to pin higher fields, one cannot increase the heavy ion irradiation dose indefinitely. It must not approach or exceed the critical fluence that destroys the superconductivity itself. Irradiation induced increase of the current density has been known [1] also for A-15 superconductors

4. Conclusion

It is concluded for HTSC oxides that the well-known [3, 6] decrease in T_c due to ion-irradiations can be observed only for the optimally-doped or underdoped samples. Typical results like a rise in T_c for (Bi, Pb)-2212 with T_{c0} of 59.5 K have always been for over-oxygenated samples. This is true for radiation damage in Bi-2212 as well as in (Bi, Pb)-2212. The irradiation induced increase or decrease of T_c and the magnitude of the change can be explained from irradiation induced change of carrier density in the HTSC. This is a change in the electronic structure of the HTSC, recently discussed in terms of the van Hove scenario.

Understanding of the direction of change in T_c due to radiation damage in BCS-type A-15 superconductors from induced change of the electronic structure is easier. The simple BCS expression for T_c predicts an increase of T_c if irradiation can increase the density of states at E_F, DOS (E_F) without adversely affecting other factors. The unirradiated T_c of common A-15 compounds like Nb_3Sn, Nb_3Ge and V_3Ga in high, compared to other conventional superconductors, due to a the peak of their DOS falling on E_F making DOS(E_F) rather high. However, for A=15 compounds like Mo_3Ge and Mo_3Si, the DOS peak or peaks just misses or miss E_F, making their T_c low compared to other superconducting A-15 compounds. Defects, generated by radiation damage, smear or somewhat flatten the sharp peak of the density of states, lowering DOS (E_F) in high-T_c A-15 compounds and raising DOS(E_F) in low-T_c A-15 compounds. This immediately explains why radiation damage can decrease T_c in the high-T_c (and hence better known) A-15 compounds, and increase T_c, at least initially, in Mo_3Ge and Mo_3Si, the low-T_c A-15 compounds. However, irradiation at still higher doses can damage significantly the overall A-15 structure and degrade T_c in all cases. Such final degradation has also been observed for Mo_3Ge.

Present overview of energetic particle radiation damage studies in so-called HTSC and conventional superconductors, outlines our understanding of the radiation induced increase of T_c in both type of superconductors from the radiation induced change of electronic structure. The same models explain the commonly observed degradation of T_c on irradiation. Present review should remove all possible doubts on increase of T_c in radiation damage under certain situations, as it identifies such situations with explanation.

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